

Summary of ARM Aerosol Working Group Report

Two of the primary objectives of ARM are: 1) relate observations of radiative fluxes and radiances to the atmospheric composition and, 2) use these relations to develop and test parameterizations to accurately predict the atmospheric radiative properties. Consequently, ARM has pursued measurement and modeling activities that attempt to determine how aerosols impact atmospheric radiative transfer, both directly and indirectly. These efforts have primarily focussed on measurements of aerosol optical thickness, retrievals of vertical profiles of aerosol scattering and extinction, and surface measurements of aerosol optical (i.e. scattering, absorption, extinction) and physical (i.e. size, composition) characteristics. However, although these efforts have provided valuable insight regarding aerosol properties and the impact of aerosols on radiation, ARM must pursue additional measurement and modeling studies to accurately address how aerosols impact radiative fluxes and radiances throughout the entire column. These studies include:

- **Aerosol absorption**
Attempts to reconcile measurements and models of clear sky diffuse radiance and flux over the SGP site have been unsuccessful due in large part by the uncertainty in the vertical profile of aerosol absorption. This uncertainty must be reduced to evaluate efforts to model diffuse radiances and fluxes.
- **Aerosol humidification and AOT closure**
Measurements of aerosol scattering and absorption and aerosol size distribution by surface and aircraft in situ instrumentation are normally referenced to a dry aerosol (relative humidity = 40%) which often does not reflect the ambient atmospheric conditions. Additional efforts should be made to determine the vertical variability of the aerosol humidification factor used to relate the dry aerosol measurements to ambient conditions and to evaluate the aerosol extinction retrieved from in situ and lidar measurements and computed from measurements of aerosol size and composition.
- **Aerosols and clouds**
Measurements of cloud condensation and ice nuclei are required to study the impacts of aerosols on cloud radiative properties and cloud lifetime.
- **Aerosol data dissemination**
Additional efforts should be made to help gather, process, and disseminate ARM aerosol data to both the ARM and outside communities.
- **Automated retrievals of aerosol properties**
Consistent with its efforts to provide long-term, continuous measurements of atmospheric composition, ARM should pursue efforts to develop and evaluate routine and automated measurements of aerosol properties aloft.

These activities require close cooperation between the aerosol measurement and modeling communities and also require the Aerosol Working Group work to work closely together with the other ARM working groups (Cloud, Shortwave, etc.).

ARM Aerosol Working Group Report, July 2000

Background

As stated in the ARM Science Plan, the ARM programmatic objectives are:

1. Relate observed radiative fluxes and radiances in the atmosphere, spectrally resolved and as a function of position and time, to the temperature and composition of the atmosphere, specifically including water vapor and clouds, and to surface properties, and sample sufficient variety of situations so as to span a wide range of climatologically relevant possibilities.
2. Develop and test parameterizations that can be used to accurately predict the radiative properties and to model the radiative interactions involving water vapor and clouds within the atmosphere, with the objective of incorporating these parameterizations into general circulation models.

One of the several key scientific issues that must be resolved in order to achieve these objectives is: "What are the direct effects of temperature and atmospheric constituents, particularly clouds, water vapor and aerosols on the radiative flow of energy through the atmosphere and across the Earth's surface?" ARM has identified the importance of defining the atmospheric state for determining atmospheric radiative transfer and, therefore, has tried to support measuring and modeling aerosols to study the impact of aerosols on radiative transfer. We shall review the key aerosol measurements and what modeling and analyses have been performed to assess aerosol impacts on radiation.

The basic parameters needed to describe how aerosols *directly* interact with solar and infrared radiation are the aerosol optical thickness (AOT), single scattering albedo ω_0 , and the phase function ϕ . The aerosol optical thickness is the integral with altitude of the aerosol extinction coefficient; the aerosol extinction coefficient is the sum of the absorption and scattering coefficients. The aerosol single scattering albedo is the ratio of the scattering to extinction coefficients. The phase function describes the angular distribution of the intensity of light scattered by the aerosols. Computation of radiative fluxes at various altitudes requires knowledge of these properties as a function of altitude. Ideally, these measurements should be acquired at several wavelengths in both the visible and infrared portions of the spectrum.

How are current ARM measurements addressing these parameters? We shall present a brief summary of the on-going activities regarding aerosol measurements and modeling as well as some recent aerosol-related studies. A more complete description of these measurement and modeling activities is given in Appendix A and a collection of slides discussing aerosol measurement and modeling activities is given in Appendix B. These summaries are based on brief presentations given at the Aerosol Working Group Meeting held on March 13, 2000 during the ARM Science Team meeting as well as an oral summary presented by Dr. Steve Schwartz during this Science Team meeting.

ARM Aerosol Measurements

The surface Aerosol Observing System at the SGP site measures aerosol light scattering and absorption coefficients, hemispheric backscattering coefficient, hemispheric backscatter fraction, single scattering albedo, Angstrom exponents, condensation nucleus concentration, and ozone. Additional measurements at SGP include the aerosol size distribution and hygroscopic growth factor. Both sets of measurements are made continuously on a daily basis in an automated manner. Since these AOS measurements are made at a low relative humidity, the hygroscopic growth factor measurements are used to relate the measurements of aerosol scattering and extinction to the values anticipated at the ambient (higher) relative humidity. Quality-checked data are available from the NOAA CMDL web site (<http://www.cmdl.noaa.gov/aero/data/>). Additional support is required to get these quality-checked data on the ARM archive.

The Cimel Sun photometer located at the SGP site, which is part of the Aerosol Robotic Network (AERONET) (Holben et al., 1998), measures column-averaged AOT on an automated basis. These data are available from both the AERONET web page (<http://aeronet.gsfc.nasa.gov:8080/>) as well as from the ARM Archive. The Multi-Filter Rotating Shadowband Radiometer (MFRSR) also measures total optical thickness, from which column-aerosol optical thickness can also be derived. Total optical thickness data for the SGP and other sites are available from the web site <http://hog.asrc.cestm.albany.edu>. Work is currently underway on Value-Added Procedure (VAPs) to produce total and aerosol optical thickness measurements that would be available from the ARM Archive. Both the Cimel Sun photometer and the MFRSR measure AOT only during daytime, cloudfree periods. The Rotating Shadowband Spectroradiometer (RSS) at the SGP measures spectrally resolved direct-normal, diffuse-horizontal, and total-horizontal irradiances using the automated shadowband technique also used for the MFRSR.

The CART Raman lidar measures profiles of aerosol extinction, aerosol backscattering, and aerosol optical thickness over the SGP site on a routine basis. The Raman lidar measurements of aerosol extinction and optical thickness (355 nm) are independent of the Sun photometer measurements. This same lidar also measures profiles of water vapor mixing ratio in the same scattering volume as the aerosol measurements. The Raman lidar measurements are available from the ARM Archive. The Micropulse Lidar (MPL) measures aerosol backscatter profiles (523 nm); using the AOT measured by the Cimel Sun photometer or MFRSR to periodically calibrate the MPL retrievals can produce profiles of aerosol extinction and AOT. Since the MPL aerosol algorithms are still under development, these MPL aerosol extinction profiles are not currently available on a routine basis.

In the In-situ Aerosol Profiling (IAP) program aerosol scattering and absorption are measured (at relative humidity = 40%) over the SGP site between 500-11,000 feet using a small aircraft. These flights, which began in March 2000 and will continue for a year, occur up to 3 or 4 times per week and are being used to obtain a statistically significant sample of vertical profiles and to compare these with the surface in-situ measurements and lidar measurements.

During the Aerosol Intensive Operations Periods (IOPs) that occurred in April 1997, September-October 1997, and August 1998, aerosol measurements were obtained using in situ instrumentation on board the Battelle Gulfstream-1 aircraft. Vertical profiles of aerosol

scattering, backscattering, and absorption extending to 5-6 km were acquired along with measurements of the aerosol size distribution. As in the IAP measurements above, the sampled air was dried as necessary so that the relative humidity was = 40%.

Present status of Aerosol Science Studies

How have these measurements been used? Several studies have attempted to look at individual aerosol optical thickness measurements and whether the various methods to retrieve AOT are consistent. Schmid et al. (1999) compared the AOT derived from 4 instruments (Cimel Sun photometer, MFRSR, RSS, and NASA Ames AATS-6 Sun Photometer) over the SGP site during the Fall, 1997 and found the AOT between 380 and 1020 nm could be retrieved with an accuracy at most wavelengths of 0.026 at the 95% confidence level compared with the WMO target of 0.02. Bergin et al. (2000) determined that the AOT over the SGP site could not be estimated using the surface in situ aerosol measurements of extinction unless information on the vertical profile of extinction is also available. When MPL profiles of normalized aerosol backscatter were used, along with hygroscopic growth factors to account for particle hygroscopicity, the AOT inferred from surface extinction values agreed within an uncertainty of 20% with the AOT measured by the MFRSR. This study also found that much (~50%) of the aerosol over the SGP site is above the mixed layer. Turner et al. (2000) examined Raman lidar aerosol extinction profile measurements acquired during 1998 and 1999 and also found that high values of aerosol extinction often occurred at levels 3-5 km above the surface. They also found that the scale height for aerosol extinction increased from about 1 km in the winter to about 2.5 km in the summer. This seasonal variability in the aerosol scale height differed from that found for water vapor; the scale height for water vapor remained at about 2 km throughout the year.

Kato et al. (2000) used aircraft measurements of scattering and extinction to examine whether the AOT determined from these instruments matches the AOT measured by Sun photometry. For those cases in April and September 1997 when the relative humidity was low, the AOT derived from the aircraft in situ measurements matched the AOT derived from Sun photometry. For the cases in August 1998 when the boundary layer relative humidity was high, and a hygroscopic growth factor correction determined from surface measurements was applied to the aircraft data, the AOT derived from the aircraft in situ measurements of aerosol scattering and extinction was about 25-31% lower than that derived from Sun photometry. The low estimate of AOT by the airborne in situ measurements of scattering and absorption was attributed in part to the 1 μm upper limit on the particle size measured by the aircraft instruments. Based on these comparisons, Kato et al. (2000) also concluded that the single scattering albedo of the aerosol particles in the lower troposphere is between 0.84 and 0.97. Liu and Daum (2000) compared aerosol scattering derived from airborne measurements of aerosol size distributions and number concentrations with aerosol scattering measurements from an airborne in situ nephelometer. They found good agreement between these values once the airborne optical particle counter were calibrated with an algorithm to account for the difference between the refractive indices of the measured and calibration aerosol particles.

Various studies have also examined how accurately radiative transfer models represent diffuse radiance in the cloud-free sky. Harrison et al. (1999) compared model radiances with RSS measurements of the spectral dependence of the direct/diffuse ratio and concluded that the

difference suggests a modest “clear sky anomaly” in absorption. The spectral signature of the diffuse “anomaly” suggests aerosol and not gaseous absorption. Harrison et al. (1999) indicated that measurements of aerosol properties above the site, in particular aerosol absorption, would be highly desirable to investigate this anomaly. Halthore and Schwartz (2000) also found that present models accurately estimate the direct beam forcing but overestimate the diffuse forcing when using measurements of AOT and “reasonable” aerosol properties. Mlawer et al. (2000) also compared direct and diffuse RSS measurements with calculations and found no evidence for unknown molecular absorption. To bring measured and modeled diffuse irradiances into agreement required single scattering albedo values of between 0.6-0.85, which are considerably lower than are generally assumed “reasonable” at the SGP site. Harrison et al. (2000) indicated that part but not all of the discrepancy might be due to incorrect assumptions regarding the amount of NO₂ over the SGP site. Assuming too small an amount of NO₂ would lead to an overestimate of AOT at shorter wavelengths; the missing NO₂ absorption may be falsely interpreted as aerosol absorption. Their analysis suggests NO₂ may be higher than commonly assumed. Additional work must be done to resolve this issue.

Studies are also underway to use a coupled climate/chemistry model to simulate the global aerosol cycle. This model, which uses emission inventories of major aerosol types, is being used to estimate aerosol direct and indirect radiative forcing (Chuang et al., 2000). The model also uses parameterizations of aerosol optical properties with relative humidity; these parameterizations vary with aerosol type and wavelength. Developing and assessing these global aerosol models requires additional data. Measurements of the aerosol size distribution, chemical composition, optical thickness, and vertical profile of optical thickness are required for modeling the aerosol direct effect, and measurements of the aerosol size distribution, cloud drop number concentration, cloud optical thickness, and cloud liquid water content profile are required for modeling the aerosol indirect effect.

Direction of Future Efforts

1. Aerosol absorption and closure of measured and modeled clear sky diffuse radiances

How do these measurements and science studies relate to the ARM science goals? As discussed above, in order to determine how aerosols directly impact radiation, the primary aerosol parameters required are the aerosol optical thickness, single scattering albedo, and backscatter phase function. Current ground based measurements made by the MFRSR, Cimel Sun photometer, and RSS are able to provide this measurement during cloud-free conditions. However, as was pointed out during summary talks above, there is considerable uncertainty in the values of aerosol absorption and single scattering albedo that have been derived from various methods. The values of ω_0 derived by the AOS tend to be highest (~0.9-1.0). The values derived from airborne in situ measurements of scattering and absorption and ground based measurements of aerosol extinction tend to be lower (0.8-0.95). Forcing modeled diffuse spectral fluxes to match spectral measurements of diffuse flux made by the RSS produced the lowest values (0.6-0.85).

There should be systematic efforts made to reconcile these differences in ω_0 . The AOS retrievals are the most direct measurements since both aerosol scattering and absorption are measured for

dry aerosol particles at the surface. However, questions were raised regarding whether these values may be biased high since there are brief periods when ω_b is reported above 1.0. Filter based measurements of aerosol absorption are perhaps questionable because of the sampling process on the filter; the shape of the particles may be changed and the interaction of the aerosols and the filter/plate system are not properly described by Beer's law (Moosmüller et al., 2000). Since filter methods also do not properly account for the influence of relative humidity on aerosol light absorption, it has been difficult to determine how aerosol absorption varies with relative humidity. Another question is whether the AOS measurements are representative of the aerosols above the columns and how the surface values compare with the column-averaged values that are derived by the other methods.

An attempt at resolving some of these aerosol absorption measurement issues was made during the aerosol absorption IOP that occurred at the SGP site during March 2000. The filter based aerosol absorption measurements were compared to the laser photoacoustic measurements. The filter based Particle Soot Absorption Photometer (PSAP), which has been the standard method for measuring absorption in the AOS system, monitors absorption by measuring transmission through a filter. The photoacoustic method measures the sound pressure produced in an acoustic resonator caused by light absorption (Arnott et al., 2000). Initial results from this experiment indicated that for dry (low RH) aerosol measurements, both instruments tracked absorption well. However, aerosol absorption measured by the PSAP was actually significantly higher than the absorption measured by the photoacoustic instrument indicating that the single scattering albedo that measured by the PSAP is likely to be biased too low rather than too high. Although the PSAP operated as part of the SGP AOS system operates at low (<40%) relative humidity, additional comparisons were performed to determine how the PSAP would operate at higher ambient relative humidity. These comparisons indicated that the PSAP aerosol measurement bias error became worse at higher relative humidity and that the PSAP absorption measurements did not track the photoacoustic measurements well at high relative humidity.

Additional uncertainty is added due to the vertical variability of aerosol properties. Initial flights associated with the IAP program have often shown large vertical variability in aerosol absorption. However, Kato et al. (2000) show that the vertical variability in ω_b is not sufficient to resolve the differences among the airborne and surface ω_b measurements and the ω_b derived from the comparisons of direct and diffuse measurements.

It is crucial to resolve the differences between these direct measurements of aerosol absorption and the indirect measurements that infer aerosol absorption by matching modeled and measured spectral diffuse measurements. We recommend that these studies of diffuse radiation concentrate on days with simultaneous measurements of ω_b acquired by both types of surface measurements discussed above as well as airborne profiles of aerosol absorption. For these measurements, the photoacoustic absorption measurements could be used to “calibrate” the surface PSAP measurements as well as a PSAP on an aircraft. The goal of these measurements would be to accurately constrain the lower limit on ω_b throughout the atmospheric profile during periods of low AOT and to then compare the measured absorption with that derived from the comparisons of modeled and measured diffuse radiation. Thus, it is crucial that accurate surface radiation measurements (both broadband and spectrally resolved) be acquired simultaneously with these aerosol measurements. We recommend that both the Aerosol and Shortwave Working

Groups closely coordinate the development and implementation of such an IOP to address these issues.

In order to help check the measurements of aerosol absorption, it will be important to also simultaneously measure aerosol scattering in order to combine both measurements to derive aerosol extinction. These measurements of aerosol scattering could be acquired by an airborne nephelometer. The resulting measurements of aerosol extinction and AOT should be compared with airborne Sun photometer and lidar measurements of AOT. The airborne Sun photometer AOT measurements would provide an important check on these retrievals. These airborne measurements should be acquired from the same aircraft since extinction or layer AOT closure between in-situ and Sun photometer measurements has been achieved only in those studies (e.g. ASTEX, TARFOX, ACE-2, INDOEX) (Clarke et al., 1996; Hegg et al., 1999; Hartley et al., 2000; Collins et al., 2000) where both measurements were taken from the same airplane. Airborne measurements of chemical composition are also desirable in order to help achieve closure between the observed and modeled direct and diffuse beams (Satheesh et al., 1999; Conant, 2000). Measurements of aerosol composition are also desired for estimating CCN as discussed below. This IOP may also provide an opportunity to deploy an airborne high spectral resolution oxygen A band spectrometer (ABS). This instrument, which is being developed by NASA Langley Research Center, is an airborne version of the ABS to be used on the Pathfinder Instrument for Cloud and Aerosol Spaceborne Observations (PICASSO) instrument, which is to begin spaceborne operations in 2003. The ABS is to be used to retrieve both aerosol and cloud optical thickness, aerosol single scatter albedo, and cirrus phase function.

There is considerable uncertainty in how models represent AOT for longer wavelengths ($>1\text{-}2\text{ }\mu\text{m}$). This uncertainty also hampers efforts to reconcile measurements and models of clear sky diffuse fluxes. Since the MFRSR and Cimel Sun photometer do not measure AOT for wavelengths longer than about $1\text{ }\mu\text{m}$, it is difficult to resolve these differences. Therefore, we recommend ARM pursue additional efforts to measure AOT beyond $1\text{ }\mu\text{m}$. Such an effort could involve the development/modification of Sun photometers to measure AOT in the near infrared.

2. Aerosol hygroscopic growth factor (closure of AOT)

Most routine aerosol measurements by in situ instruments dry the aerosols to a low relative humidity before measuring the physical (size, shape, composition) and optical (scattering, absorption, phase function) properties. The airflow is generally heated slightly (if necessary) so that the aerosol properties are measured at a reference relative humidity of around 40%. This is done because it is typically very difficult to accurately measure aerosol properties at the ambient relative humidity. These measurements of dry aerosol properties also make it easier to relate aerosol physical and optical characteristics to aerosol inventories used in simulating aerosol cycles in global numerical models. However, since many aerosols are hygroscopic, the size and scattering properties of aerosols typically are highly dependent on high relative humidity. Therefore, it is crucial to have accurate parameterizations of how the aerosol physical and optical characteristics vary with RH. Presently, there are routine measurements of the aerosol hygroscopic growth factor (or humidification factor) (i.e. variation in scattering coefficient with RH) made by the AOS at the surface at the SGP. Since few measurements have been made to determine how aerosol absorption varies with RH, parameterizations of how aerosol absorption

are highly uncertain. In the case of highly absorbing aerosols, this uncertainty also impacts the parameterizations of how aerosol extinction varies with RH.

Unfortunately, the measurements discussed above have only occurred at the surface; there is very little information on how the aerosol hygroscopic growth factors vary with altitude. While some limited results derived from the Raman lidar suggest that this factor may be slightly greater at higher altitudes than the values derived from the AOS surface measurements, the uncertainty of these lidar measurements, as well as the different wavelengths between the lidar and AOS measurements, make it difficult to characterize the altitude dependence of the aerosol hygroscopic factor. Since the IAP profile measurements of scattering and absorption (and consequently extinction) are for the dry aerosols, the uncertainty in how the humidification factor varies with altitude will impact comparisons of aerosol extinction profiles measured by the lidars and derived from IAP measurements.

Therefore, we recommend measuring the aerosol hygroscopic growth factor as a function of altitude. These measurements would be used to convert the measurements of dry aerosol scattering to ambient conditions and to test closure for retrieving aerosol scattering and extinction from in situ aerosol measurements. These measurements would also be used to determine if the surface measurements of the aerosol hygroscopic factor can be used to estimate this factor for the vertical profile. In addition to using both “dry” and “wet” nephelometers to measure this humidification factor, an additional nephelometer that measures aerosol scattering at ambient RH (or as close to ambient as possible) would be desirable. The aircraft measurements should also include collection of aerosol samples for additional analyses of chemical composition as well as measurements of the aerosol size distribution. The results of the chemical composition analyses could be used along with the measurements of the aerosol humidification factor to develop and assess parameterizations of the humidification factors used in aerosol models.

As in the case of aerosol absorption discussed in item 1, it will be important to compute aerosol scattering, absorption, and extinction during high relative humidity conditions ($RH > 70\%$) and compare the resulting estimate of aerosol extinction and AOT with those derived from lidar and airborne Sun photometer measurements.

3. Aerosols and Clouds

The studies above address the direct effects of aerosols on radiation. Aerosols also impact atmospheric radiation indirectly by affecting cloud properties. Aerosols may increase cloud reflectivity due to more and smaller cloud droplets forming on the aerosol (“Twomey” effect), and by increasing the lifetime of clouds due to reduced precipitation in clouds with more and smaller droplets (“Albrecht” effect) (Raes et al. 2000). The effect on cloud albedo of more numerous droplets has been described by Han et al. (1994) and the effect on precipitation and hence cloud extent has recently been indicated by Rosenfeld (2000). From in situ measurements in Florida (small cumulus clouds) and the eastern Atlantic (stratus clouds), a strong effect of higher pre-cloud particle concentrations (cloud condensation nuclei CCN) on precipitation initiation (an order of magnitude fewer drizzle drops) has been found. Even modest differences

in droplet concentrations (a factor of 2 and thus differences in droplet sizes) had profound effects on precipitation initiation, which can affect cloud extent.

To meet the ARM objectives of relating observed atmospheric radiative fluxes and radiances to clouds, both of these effects need to be quantified. ARM now has the capability of remotely measuring vertical profiles of cloud liquid water content, droplet number concentration, and droplet effective radius. But one crucial measurement is lacking: the CCN spectrum at cloud base. Since most of the presently available data has been obtained in cleaner (maritime) areas the addition of data from more polluted areas (i.e. Oklahoma) would be a large step forward for the indirect aerosol effect. ARM funded CCN spectrum measurements from aircraft during the 1997 Fall IOP, but unfortunately during that IOP there were few clouds that satisfied the requirements for remote sensing of the cloud microphysical properties, and aircraft measurements of CCN spectra were not available for any one them. Without coincident measurements of CCN spectrum and cloud microphysics it is impossible to evaluate models of the influence of aerosols on cloud microphysics. At least one additional campaign of CCN spectrum measurements is needed to go along with the excellent cloud measurements.

We also recommend that these CCN studies include direct measurements of CCN using thermal diffusion chamber(s) as well as measurements of the aerosol nucleation mode size distribution and aerosol compositions. The direct CCN measurements are important to for determining the feasibility and uncertainty in estimating CCN using nucleation mode aerosol size distributions and particle composition. An International CCN Workshop that will be held this summer to review these measurements should provide guidance regarding the appropriate instruments and techniques.

The addition of surface CCN measurements, at least during an Cloud/Aerosol IOP, would permit the evaluation of the vertical variability of CCN and would provide data to assess of the utility of continuous surface CCN measurements.

We also support the addition of high gain narrow field of view (NFOV) nitrogen Raman and Raman liquid water channels to the SGP Raman lidar. Although the main motivation for the NFOV nitrogen Raman channel would be to aid in the retrieval of cloud extinction profiles, this channel would also greatly improve retrievals of stratospheric aerosol extinction profiles. Aerosol extinction profiles derived from the current Raman lidar currently do not extend above about 12-15 km due primarily to the low signal/noise ratio in the Raman nitrogen channel. Because stratospheric aerosol loadings have been at negligibly low background levels for the past 5 years, there has been little need for accurate measurements of stratosphere extinction. However, since major volcanic eruptions have generally occurred about once per decade over the last 40-50 years, one can not expect stratospheric aerosols to remain at low levels indefinitely. Following the eruptions of the El Chichon (1982) and Pinatubo (1991) volcanoes, AOT due to stratospheric aerosols was often as large or larger than the AOT due to tropospheric aerosols. Following these volcanic eruptions, Raman lidars were to measure stratospheric aerosol extinction and optical thickness profiles as well as to characterize the size and composition of these volcanic aerosols (Ferrare et al., 1991; Ansmann et al., Wandinger et al.,).

The recently demonstrated experimental capability of the Raman liquid water vapor channel permits the liquid water content, average droplet radius and droplet number density to be determined, as a profile, in the lowest 300-500 m of a liquid cloud (Whiteman et al., 1999). Given the difficulty that the radar has in sensing the small particles that are typically present at the base of clouds, the inclusion of these Raman lidar could allow fundamental studies of cloud physics processes directly related to the indirect effect of aerosol, i.e. cloud droplet growth, to be performed in a manner not currently possible. These measurements could either be added to the CART Raman lidar or could be made available through a deployment of the NASA Scanning Raman Lidar to the CART SGP site during an Aerosol/Cloud IOP.

4. Aerosol Data Dissemination

Aerosol related data are presently acquired from a variety of sensors. A partial list of these aerosol related products include: aerosol optical thickness (AOT) (MFRSR, Cimel Sun photometer); Angstrom exponent (MFRSR, Cimel Sun Photometer, AOS), aerosol scattering and absorption coefficients (AOS (routine) and aircraft (IOP)), aerosol size distributions (AOS (routine) and aircraft (IOP)), aircraft backscatter/extinction vertical profiles (MPL, Raman lidar, aircraft (IOP)), etc. These data are presently scattered among many separate files from various data sources and are not available from a single location nor are they available as a single data product. Presently, the only data measured in a somewhat continuous manner that are currently available from the ARM Archive are: Raman lidar best estimate data (water vapor, aerosol, temperature profiles), normalized MPL backscatter profiles, Cimel Sun photometer AOT data, and raw (not quality checked) AOS data. Additional work is required to derive AOT from the MFRSR observations from the extended facilities at the SGP and to reference the AOT to the AOT obtained at the SGP Central Facility as well as to quality check and archive the AOS and profile measurements at the CF. It would greatly aid both the ARM community and outside users if these aerosol data were combined into value-added product(s) (VAPs) to facilitate the distribution and analyses of these data. Since these aerosol data are acquired from several different sensors at different temporal resolutions, the AWG recognizes the difficulty in developing a single product. For this reason, and because aerosol data acquired during IOPs are normally most often used, it is recommended that the development of an Aerosol VAP begin with the data acquired during the Spring 2000 Cloud/Arese II/Aerosol Absorption IOP that occurred in February-April 2000. An Aerosol VAP develop during this period would also be helpful for the NASA Terra Validation activities that also occurred over the SGP site during this time.

5. Automated retrievals of Aerosol Properties

Dr. Oleg Dubovik (SSAI/NASA/GSFC) has developed an inversion code that uses measurements of sky radiance and aerosol optical thickness to derive the aerosol volume size distribution including the volume median and effective radii (Dubovik et al., 2000). In addition, for $AOT > 0.4$, these routines also derive the aerosol refractive index, aerosol single scattering albedo, and phase function. Details of these inversions may be found at <http://aeronet.gsfc.nasa.gov:8080/>. The column-averaged aerosol size distribution information derived from this inversion may provide a useful supplement to the surface-based size distributions derived from the AOS since the aerosol properties derived from the inversion are

for the entire atmospheric column instead of just at the surface. These retrievals represent the only routine, automated method of retrieving aerosol size distribution information for the atmospheric column. Therefore, these retrievals can potentially provide a climatological database of aerosol size distributions that can be used by the ARM program. These data could be used to evaluate how well the surface based aerosol size distributions represent the entire column. In addition, since there are many Cimel Sun photometers throughout the world as part of the AERONET program, data from these instruments can potentially be used to retrieve similar size distribution information for different aerosol types throughout the world. Data from the AERONET program will be used extensively to validate aerosol properties retrieved from the Terra MODIS instrument.

It is important to compare the aerosol size distribution (and refractive index and single scattering albedo, when applicable) derived from the Cimel retrieval algorithms with aerosol size distributions measured as a function of altitude by aircraft in situ sensors in order to determine the capabilities and limitations of these aerosol retrievals. For the next Aerosol IOP, we recommend using an airborne optical particle counter to measure the aerosol size distribution simultaneously with the Cimel Sun photometer measurements of sky radiance and AOT. Since the airborne in situ aerosol measurements are acquired at low RH, it is important to acquire measurements during both low and high ambient RH in order to determine the impact of RH corrections on the in situ aerosol size distributions.

A new method to remotely evaluate the vertical variability of effective particle radius, volume and surface concentrations, refractive indices and single scattering albedo within an atmospheric column has been discussed by Ansmann et al. (2000) and Müller et al. (2000). This method uses an unique six-wavelength aerosol lidar to measure aerosol backscattering at these wavelengths as well as aerosol extinction at two of these wavelengths. From these aerosol backscattering and extinction profiles, an inversion algorithm is used to derive the aerosol parameters listed above. This lidar system was recently used to characterize the vertical variability of aerosols over the Maldives Island during the Indian Ocean Experiment (INDOEX). We encourage ARM to investigate the use of this system during an IOP to determine the feasibility of using this technique to measure the vertical variability of aerosols.

6. Aerosol Impact on Shortwave Flux

Attempts to reconcile measurements and calculations of shortwave diffuse flux are limited by measurements of shortwave flux as well as measurements of AOT. Diffuse flux measurements by the shaded Eppley Precision Spectral Pyranometer (PSP) require a correction to account for the effect of the dome-detector thermal IR transfer. An attempt to correct for this using the net IR signal measured by the pyrgeometer has been only partially successful. Another method uses the thermistors mounted on the dome of the PSP to monitor temperature gradients within the instrument. Preliminary results of a correction scheme using these thermistor temperature measurements have shown that the PSP correction is much larger and more variable than the previous first order correction. These results indicate the need for implementing a thermistor-based correction scheme on routine basis. An alternative method of measuring diffuse fluxes would use Eppley Black and White (B&W) Shortwave pyranometers. These instruments do not have the thermal offset problems associated with the PSPs. The NOAA Surface Radiation

Research Branch (SRRB) has decided to use the Eppley (B&W) pyranometers to measure diffuse SW irradiance as part of the Surface Radiation Budget Network. During the Spring IOP at the SGP, both methods of measuring diffuse flux are being tested. These tests should include comparisons of diffuse radiation measured under conditions of clear, thin, and optically thick clouds to evaluate possible directional effects of diffuse radiation measurements. We urge ARM to employ one (or both) of these methods for routine measurements of diffuse flux.

General Recommendations

In order to pursue these activities successfully, it is crucial that ARM and the Aerosol Working Group develop specific science team(s) to address these issues. These team(s) would work out the specific science objectives, determine and prioritize the key measurements, identify the appropriate instruments, and carry out the appropriate measurement and modeling activities. The science team(s) would also be responsible for communication/presentation/publication of these results, perhaps including combining the publication of these results in a journal special issue. It is important that these science teams be comprised of personnel from both measurement and modeling groups. Since these activities involve issues addressed by other ARM working groups (e.g. shortwave, cloud, instantaneous radiative flux, etc.), these teams should involve key personnel from these other working groups.

ARM can support these activities by placing high priority on science studies to address these activities. These science studies should focus on the use and analyses of existing data, including both routine and IOP measurements, as well as on the acquisition and analyses of additional data. ARM can also support these activities by providing additional infrastructure resources to help with the collection and dissemination of data. The extensive list of additional measurements required to address these science issues discussed above will require resources beyond those that have already been deployed for previous Aerosol IOPs. These additional resources would be needed for both surface and aircraft measurements. ARM and the Aerosol Working Group must work together with outside agencies to successfully carry out these experiments.

- **Recommended measurements for future Aerosol/Cloud/Shortwave IOP**

1. AOS measurements
 - a. aerosol light scattering at 3 wavelengths
 - b. aerosol absorption coefficients (filter+photoacoustic methods)
 - c. hemispheric backscattering coefficient and hemispheric backscatter fraction
 - d. single scattering albedo
 - e. Angstrom exponents
 - f. condensation nucleus concentration
 - g. ozone.
 - h. aerosol size distribution
 - i. scattering and absorption hygroscopic growth factors
2. Aircraft measurements
 - a. aerosol size distribution
 - b. aerosol scattering at 3 wavelengths
 - c. aerosol absorption coefficient (multiple wavelengths if possible)
 - d. hemispheric backscattering coefficient and hemispheric backscatter fraction
 - e. single scattering albedo
 - f. Angstrom exponents
 - g. condensation nucleus concentration
 - h. scattering (and if possible absorption) growth factors
3. Cimel Sun photometer
 - a. AOT
 - b. Sky radiance to derive
 1. aerosol size distribution
 2. refractive index and single scattering albedo if possible
4. MFRSR
5. RSS
 - a. direct spectral irradiance
 - b. diffuse spectral irradiance
6. CART Raman lidar
 - a. water vapor and relative humidity profiles
 - b. aerosol backscattering and extinction profiles
7. MPL
 - a. aerosol profiles
8. Airborne Sun photometer
 - a. profiles of AOT/aerosol extinction (These measurements are desired since Sun photometry has demonstrated AOT accuracy within 0.01-0.02. AOT profiles from airborne Sun photometer would provide the baseline to assess other methods (airborne in situ scattering and extinction, computed from airborne size distribution, lidar, etc.)
9. Broadband Shortwave Diffuse Flux
 - a. shaded Eppley PSP with dome-mounted thermistor
 - b. Eppley B&W pyranometer
10. ASD Spectroradiometer
 - a. AOT for wavelengths beyond 1 μm
11. Tower mounted nephelometer
 - a. aerosol scattering at 60 m
12. six channel advanced aerosol lidar
 - a. aerosol backscattering and extinction profiles
 - b. aerosol volume and surface concentration profiles
 - c. aerosol refractive index profiles
 - d. aerosol single scattering albedo profiles
13. high resolution oxygen A band spectrometer
 - a. aerosol and cloud optical thickness
 - b. aerosol single scatter albedo

References

- Ansmann, A., U. Wandinger, and C. Weitkamp, One-year observations of Mount-Pinatubo aerosol with an advanced Raman lidar over Germany at 53.3 N, *Geophys. Res. Letters*, 20, 711-714, 1993.
- Ansmann, A., D. Althausen, U. Wandiger, K. Franke, D. Müller, F. Wagner, and J. Heintzenberg, Vertical profiling of the Indian aerosol plume with six-wavelength lidar during INDOEX: A first case study, *Geophys. Res. Letters*, 27, 963-966, 2000.
- Arnott, W.P., H. Moosmüller, C. F. Rogers, T. Jin, and R. Bruch, Photoacoustic spectrometer for measuring light absorption by aerosol: Instrument description, submitted to *Atmos. Environment*, 2000.
- Bergin, M.H., S.E. Schwartz, R.N. Halthore, J.A. Ogren, and D.L. Hlavka, Comparison of aerosol optical depth inferred from surface measurements with that determined by Sun photometry for cloud-free conditions at a continental U.S. site, *J. Geophys. Res.*, 105, 6807-6816, 2000.
- Chuang, C. C., J. E. Penner, K. E. Grant, Prospero, J. M. and G. H. Rau, Effects of anthropogenic aerosols on cloud susceptibility: A sensitivity study of radiative forcing to aerosol characteristics and global concentration. Submitted to *J. Geophys. Res.*, 2000.
- Clarke, A. D., Porter, J. N., Valero, F. P. J., and P. Pilweskie, Vertical profiles, aerosol microphysics, and optical closure during the Atlantic Stratocumulus Transition Experiment: Measured and modeled column optical properties. *J. Geophys. Res.* 101, 4443-4453. 1996.
- Collins, D. R., H. H. Jonsson, J. H. Seinfeld, R.C. Flagan, S. Gassó, D. A. Hegg, B. Schmid, P. B. Russell, J. M. Livingston, E. Öström, K. J. Noone, L. M. Russell, and J. P. Putaud, In situ aerosol size distributions and clear column radiative closure during ACE-2. *Tellus, B* 52, 498-525, 2000.
- Conant, W.C., 2000: An Observational Approach for Determining Aerosol Surface Radiative Forcing: Results from the First Field Phase of INDOEX, *Journal of Geophysical Research*, 105, 15347-15360.
- Dubovik, O., A. Smirnov, B.N. Holben, M.D. King, Y.J. Kaufman, T.F. Eck, and I. Slutsker, Accuracy assessments of aerosol optical properties retrieved from AERONET Sun and sky-radiance measurements", *J. Geophys. Res.*, 105, 9791-9806, 2000.
- Ferrare, R.A., S.H. Melfi, D.N. Whiteman, K.D. Evans, Raman Lidar Measurements of Pinatubo Aerosols over Southeastern Kansas During November-December 1991, *Geophys. Res. Letters*, 19, 1599-1602, 1992.

- Halothore, R.N. and S.E. Schwartz, Comparison of Model Estimated and Measured Diffuse Downward Surface Irradiance in Cloud-free Skies, submitted to J. Geophys. Res., 1999.
- Han, Q., Rossow, W.B., and A.A. Lacis, Near-global survey of effective droplet radii in liquid water clouds using ISCCP data. J. Climate 7, 465-497, 1994.
- Harrison, L., M. Beauharnois, J. Berndt, P. Kiedron, J. Michalsky, and Q. Min, The Rotating Shadowband Radiometer (RSS) at SGP, Geophys. Res. Letters, 26, No. 12, 1715-1718, 1999.
- Harrison, L., J. Berndt, P. Kiedron, J. Michalsky, Q. Min, and J. Schlemmer, Clear-Sky Observations from the Rotating Shadowband Spectroradiometer (RSS) at SGP, 10th ARM Science Team Meeting, San Antonio, Texas, March, 2000.
- Hartley, W. S., Hobbs, P. V., Ross, J. L., Russell, P. B., and Livingston, J. M. Properties of aerosols aloft relevant to direct radiative forcing off the mid-Atlantic coast of the United States. J. Geophys. Res., Vol. 105 , No. D8 , 9859-9886 (2000).
- Hegg, D. A., Livingston, J. M., Hobbs, P. V., Novakov, T. and Russell, P. B. 1997. Chemical apportionment of aerosol column optical depth off the mid-Atlantic coast of the United States. J. Geophys. Res., 102, 5,293-25,303.
- Holben B.N., T.F.Eck, I.Slutsker, D.Tanre, J.P.Buis, A.Setzer, E.Vermote, J.A.Reagan, Y.Kaufman, T.Nakajima, F.Lavenu, I.Jankowiak, and A.Smirnov, AERONET - A federated instrument network and data archive for aerosol characterization, Rem. Sens. Environ., 66, 1-16, 1998.
- Kato, S., M.H. Bergin, T.P. Ackerman, T.P. Charlock, E.E. Clothiaux, R.A. Ferrare, R.N. Halothore, N. Laulainen, G.G. Mace, J. Michalsky, and D.D. Turner, A comparison of the aerosol optical thickness derived from ground-based and airborne measurements, J. Geophys. Res., 105, 14701-14717, 2000.
- Liu, Y. and P. H. Daum, J. Aerosol Sci., in press, (2000).
- Mlawer, E.J., P. D. Brown, S.A. Clough, L.C. Harrison, J.J. Michalsky, P.W. Kiedron, T. Shippert, Comparison of spectral direct and diffuse solar irradiance measurements and calculations for cloud-free conditions, submitted to Geophys. Res. Letters, (2000)
- Moosmüller, H., W.P. Arnott, C.F. Rogers, J.C. Chow, C.A. Frazier, L. E. Sherman, and D.L. Dietrich, Photoacoustic and filter measurements related to aerosol light absorption during the Northern Front Range Air Quality Study (Colorado 1996/1997), submitted to J. Geophys. Res., 2000.
- Müller, D., F. Wagner, D. Althausen, U. Wandinger, and A. Ansmann, Physical properties of the Indian aerosol plume derived from six-wavelength lidar observations on 25 March 1999 of the Indian Ocean Experiment, Geophys. Res. Letters, 27, 1403-1406, 2000.

- Raes, F. T. Bates, F. McGovern, and M. van Liedekerke, The 2nd Aerosol Characterization Experiment (ACE-2): general overview and results, *Tellus*, 52B, 111-125.
- Rosenfeld, D., TRMM observed first direct evidence of smoke from forest fires inhibiting rainfall. *Geophys. Res. Let.* 26, 3105-3108, 1999.
- Satheesh, S.K., V. Ramanathan, X. Li-Jones, J.M. Lobert, I.A. Podgorny, J.M. Prospero, B.N. Holben and N.G. Loeb, 1999: A Model for the Natural and Anthropogenic Aerosols over the Tropical Indian Ocean Derived from INDOEX Data. *J. Geophys. Res.-Atmos.*, 104, D22, 27,421-27,440.
- Schmid, B., J. M. Livingston, P. B. Russell, P. A. Durkee, H. H. Jonsson, D. R. Collins, R. C. Flagan, J. H. Seinfeld, S. Gassó, D. A. Hegg, E. Öström, K. J. Noone, E. J. Welton, K. J. Voss, H. R. Gordon, P. Formenti, and M. O. Andreae, Clear sky closure studies of lower tropospheric aerosol and water vapor during ACE 2 using airborne sunphotometer, airborne in-situ, space-borne, and ground-based measurements, *Tellus*, B 52, 568-593, 2000.
- Turner, D.D., R.A. Ferrare, L.A. Heilman, W. Feltz, and T. Tooman, Automated retrievals of aerosol extinction and backscatter coefficient profiles from a Raman lidar, submitted to *J. Appl. Meteor.*, 2000.
- Wandinger, U., A. Ansmann, J. Reichardt, and T. Deschler, Determination of stratospheric-aerosol microphysical properties from independent extinction and backscattering measurements from a Raman lidar, *Appl. Optics*, 34, 8315-8329, 1995.
- Whiteman, D. N., S. H. Melfi, 1999: Cloud liquid water, mean droplet radius and number density measurements using a Raman lidar, *J. Geophys. Res.*, Vol 104 No. D24 December 27, 31411-31419.